

ATMOSPHERIC AEROSOLS

- Health hazard ($PM_{2.5}$)
 - Visibility, haze (optical depth)
I.e., light extinction
 - Climate (optical properties)
- AIR QUALITY**
CLIMATE
Direct effect
Semi-direct effect
Indirect effect
- DEPENDENT ON**
-Composition, Size
-RH, i.e., water partitioning
(related to composition, and affects size)

Aerosol Optical Properties (AOP)

- Extinction (σ_{ext}), scattering (σ_{scat}), absorption (σ_{abs}) $\rightarrow \sigma_{\text{ext}} = \sigma_{\text{scat}} + \sigma_{\text{abs}}$
- Single Scattering albedo ($\omega = \sigma_{\text{scat}} / \sigma_{\text{ext}}$) \rightarrow low ω = optically absorbing aerosols

FACT = Particle can grow upon water uptake when RH increases (hygroscopic behavior)
RESULT = Change in composition, visibility (air quality), increase of σ_{ext} (climate)

- $f_{\text{scat}}(\text{RH})$ and γ - RH dependence of aerosol extinction $\rightarrow f_{\text{scat}}(\text{RH}) = \frac{\sigma_{\text{scat}}(\text{RH})}{\sigma_{\text{scat}}(\text{Ref})}$

$$f_{\text{scat}}(\text{RH}) = \left(\frac{100 - \text{Ref } \gamma}{100 - \text{RH}} \right)^2$$

$\gamma < 0.3$, dust and black carbon
 $\gamma \sim 0.6 - 0.8$, sulphates (age)
 $\gamma \sim 0.9$, 'clean' marine
 $\gamma = 1.0 - 1.2$, H_2SO_4

INCREASING HYGROSCOPICITY
(RELATED TO SIZE, COMPOSITION)

% single term parameterization of $f(\text{RH})$
typically between 0.1 (not hygroscopic) and 1.0 (very hygroscopic)

TEXAQS - GOMACCS CAMPAIGN (July 27-September 11, 2006)

AEROSOL-RELATED SCIENCE QUESTIONS

- What are the aerosol sources in the study area?
- What are the optical and chemical properties of the aerosols?
- How do dynamics (re-circulation, land/sea breeze) and transport of polluted air affect air quality and shape the radiative budget?
- What is the direct effect of the sampled aerosols on climate?
- Are industries (typically gaseous emissions) also associated with particle emission formation - potentially additional health hazard

Let's try answering some

Cavity Ring-Down Aerosol Extinction Spectrometer (CRD-AES) on the RHB

CRD-AES in the PMEL AeroVAN



- Aerosols from common inlet (60% RH, 30 μm)
- Coupled w/ Photo Acoustic (PAS)₂₃ to get dry
single scattering albedo (ω) @ 532 nm
- Configured for measuring $f_{\text{scat}}(\text{RH})$ and γ
- Sub 1 and sub 10 μm

CRD-AES technique⁽¹⁾

Extinction = scattering + absorption (particles and gases)

N_2 purge flow

Sample (L) Cavity (L) Purge (L)

$\sigma_{\text{ext}} = \frac{L_0}{L_c} \left(\frac{1}{\tau} - \frac{1}{\tau_0} \right)$ (10¹⁰ m², or Mm⁻¹)

τ = time constant, c = speed of light

τ_0 = w/out sample

Small uncertainty ($\Delta\sigma_{\text{ext}} < 2\%$) w/ proper time resolution

MEASURED CRD-AES PARAMETERS

- σ_{ext} measured in 6 independent cavities (flow 1.5 - 3 lpm)
- σ_{ext} 532 nm @ 85%, 65%, 25% RH \rightarrow RH₅₃₂ (85,25), (65,25), and γ
- σ_{ext} 355 nm @ 75 % and 25% RH \rightarrow RH₃₅₅ (75,25)
- σ_{ext} 1064 nm @ 75 % and 25% RH \rightarrow RH₁₀₆₄ (75,25)
- ω from CRD and PAS combination, 532 nm, dry (25%RH) $\rightarrow \omega = (\sigma_{\text{scat}} / \sigma_{\text{ext}}) / \sigma_{\text{ext}}$
- Ångström exponent, Λ (dependence of σ_{ext}) $\rightarrow \Lambda = -\log(\sigma_{\text{ext},1} / \sigma_{\text{ext},2}) / \log(\lambda_1 / \lambda_2)$ $\lambda_1 = 355$, $\lambda_2 = 1064$

AOP summary for the presented aerosol types

	σ_{ext} (2.2 Mm ⁻¹) 532 nm, fine, dry	γ (0.05) 532 nm fine	ω (0.05) 532 nm fine, dry	Λ (25.1) 355, 1064 nm fine, dry
1) Ship plumes	50-100 (ship type, dilution?)	0.1-0.4	0.2-0.3	-1.5
2) Traffic	~100-150	~0.3	~0.3	~1.5
3) Continental fresh	30	~0.45-0.6	~0.7-0.8	~2.2-2.3
4) Continental processed	>50	~0.65-0.8	≥ 0.9	~2.2-2.5
5) Marine	<10	0.85-0.9	>0.97	~2.0

Future Work

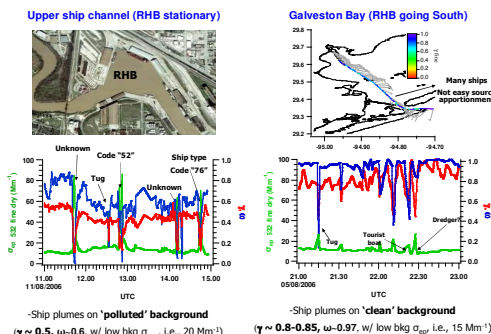
- *Continue AOP data interpretation (complexity due to multiple sources)
- *Characterization of air masses (e.g., FLEXPART for air mass trajectories)
- *Look further at interesting happenings (e.g., $\gamma > 1$ in the proximity of SO₂ sources)

The AOP dependence on RH (RH) needs to be included in both air quality (PM_{2.5} and visibility forecasting) and radiative forcing models (aerosol direct effect).....use γ values as accurate estimate of $f(\text{RH})$

HOW DO THE OPTICAL PROPERTIES OF THE MEASURED AEROSOLS DEPEND ON COMPOSITION (SOURCES), TRANSPORT, METEOROLOGY AND TRANSFORMATION ?? - SOME CASE STUDIES

Aerosol emissions from commercial vessels

Hundreds of vessels of different types (AIS database, see Eric Williams)

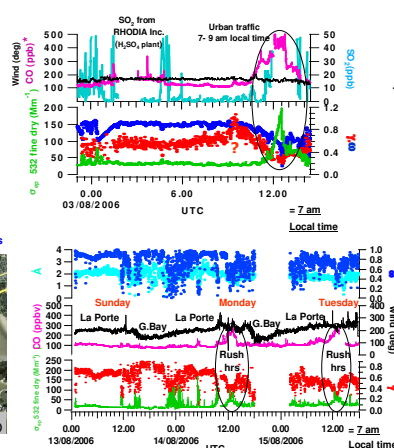
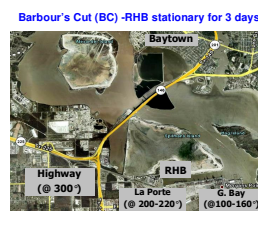
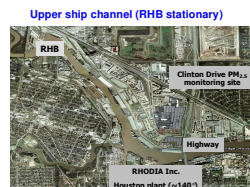


-Ship plumes on 'polluted' background ($\gamma \sim 0.5$, $\omega \sim 0.6$, w/ low bkg σ_{ext} , i.e., 20 Mm⁻¹)
-Ship plumes on 'clean' background ($\gamma \sim 0.8-0.85$, $\omega \sim 0.97$, w/ low bkg σ_{ext} , i.e., 15 Mm⁻¹)
-Ship plumes consist of non-hygroscopic ($\gamma = 0.1-0.3$), light-absorbing ($\omega = 0.2-0.3$) particles (σ_{ext} level proportional to the number of particles emitted)
-AOP and σ_{ext} level vary (plume dilution, background or ship type/speed?)

The high number of commercial ships in the Houston/Galveston area can significantly impact both air quality and climate (high number of "black" carbon particles)

Urban plumes (morning traffic rush hrs)

Detected in several occasions in the Houston area, and Beaumont (not shown) - repeatable signature, well recognizable among other sources



- Urban plumes AOPs
- Low γ (~ 0.3), low ω (0.2-0.3), $\Lambda = 1.7$ (see below)
(non hygroscopic, optically absorbing, quite small particles)
correlated with high σ_{ext} levels (150 Mm⁻¹)
- "Fresh" plume (high CO and toluene/benzene) - 3.5 - not shown
* CO from B. Lemmer, ** VOCs from J. Gilman, P. Goldan, W. Kuster
- ? = broad increase in $\gamma \sim 1.1$ (typical γ for H_2SO_4)
Oxidation of SO₂ to H₂SO_{4(aq)} (via radical chemistry?) and condensation into pre-existing aerosol particles?
- Could particles also be directly emitted from Rhodia Inc.?
(see σ_{ext} and SO₂ @ 00 UTC)
- As above, traffic plumes show $\gamma = 0.3$, $\omega = 0.3$, $\Lambda = 1.6$
(σ_{ext} increase detected @ 1064 nm, likely road dust)
- Difference between weekend and weekday mornings
(traffic signature missing on Sunday mornings)
- Air masses from many surrounding areas, following a repetitive pattern (typical daily sea-land breeze shift)
- Many other aerosol sources in BC (e.g., high σ_{ext} @ 15 UTC on 13/08 and 00 UTC on 14/08, w/ $\gamma = 0.3$, $\omega = 0.2$, $\Lambda = 1$ along w/ high SO₂ and sulphate)

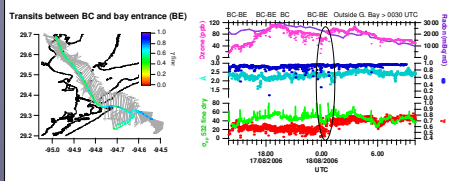
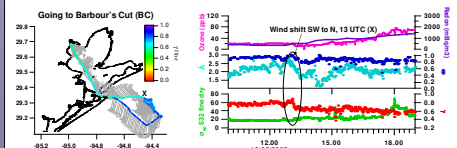
Continental Outflow and Pollution Events During TeXAQS - GoMACCS

Events selected based on high background σ_{ext} levels (532 fine dry > 50 Mm⁻¹ for several hrs), indicating photochemical transformation and processing of primary emitted aerosols

AEROSOL SOURCES - Pollution events associated w/ Northerly flow (Houston area/inland Texas, mostly anthropogenic sources). Aerosol composition (PMEL) shows rapid increases in organics (POM) and non sea salt sulphate (nss SO₄²⁻)

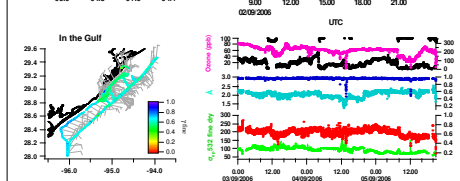
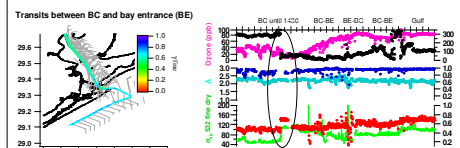
IMPLICATIONS for AIR QUALITY - development of toxic compounds (i.e., photochemical smog), visibility impairment (haze) and CLIMATE - oxidized aerosols are more hygroscopic, scatter more light (> ω). Change in radiative properties = MAJOR CLIMATE ISSUE

Galveston Bay - Aug. 16, 17, 18

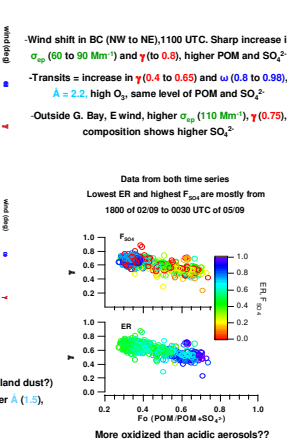


- During transits = decrease in γ (0.6 to 0.5), but increase in ω (0.9 to 0.95), and Λ (2.1 to 2.3)
Wind NE/E (see radon trend) carrying POM, SO₄²⁻ (same levels)
- Outside G. Bay, wind shift at 00 UTC, sharp increase in γ (0.7 to 0.7), $\Lambda = 2.5$, $\omega = 0.96$
- SO₄²⁻ - POM. Too low Na⁺ (< 0.2 $\mu\text{g}/\text{m}^3$) and too high σ_{ext} level for marine air
- Possibly a slightly processed air mass on Aug. 18 from 00 to 06 UTC

Galveston Bay/Gulf - Sept. 2-9, highest aerosol levels (> 100 Mm⁻¹ for many days, no coarse mode)



- Wind shift in BC (NW to NE), 1100 UTC. Sharp increase in σ_{ext} (60 to 90 Mm⁻¹) and γ (to 0.8), higher POM and SO₄²⁻
- Transits = increase in γ (0.4 to 0.65) and ω (0.8 to 0.98), $\Lambda = 2.2$, high O₃, same level of POM and SO₄²⁻
- Outside G. Bay, E wind, higher σ_{ext} (110 Mm⁻¹), γ (0.75), composition shows higher SO₄²⁻
- During 03-05/09 = $\sigma_{\text{ext}} > 100 \text{ Mm}^{-1}$, $\gamma = 0.8$, $\omega = 0.98$, $\Lambda = 2.2$ (high sub 1 μm 1064 nm, small size land dust?)
- Plumes 12-14 UTC on 03/09 (Freeport) and on 05/09 (Galveston) associated with high CO, lower Λ (1.3), low ω (0.4), low γ (0.4), toluene/benzene < 2 - processed urban outflow
- Likely processed air masses most of the time - hazy on 04/09



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